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Title: Negative Absolute Temperatures: 'Hot' Spins in Spontaneous Magnetic Order

Year: 1994

Version: Pre-print

Please cite the original version:

Hakonen, Pertti J. & Lounasmaa, Olli V. 1994. Negative Absolute Temperatures: 'Hot' Spins in Spontaneous Magnetic Order. Science. Volume 265, Issue 5180. 1-29. DOI: 10.1126/science.265.5180.1821

Negative Absolute Temperatures: 'Hot' Spins in Spontaneous Magnetic Order

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Depending on the sign of the spin temperature, $\pm T$, in the picokelvin range, antiferro- and ferromagnetic nuclear order in silver are caused by the same interactions. In rhodium, the antiferromagnetic state is preferred both at $T > 0$ and at $T < 0$. The lowest and 'highest' temperatures ever produced and measured, 280 pK and − 750 pK, have been reached in the course of these experiments. The results on silver, in particular, show that negative temperatures are real, not fictitious quantities.

Nuclear spins in metals provide good models to investigate magnetism. The nuclei are well localized, their spins are isolated from the electronic and lattice degrees of freedom at low temperatures, and the interactions between nuclear spins can be calculated from first principles. Therefore, these systems are particularly suitable for testing theory against experiments. Furthermore, many of the results obtained in nuclear systems do not have any counterpart in the realm of ordinary electronic magnetism.

Nuclear moments of every element with non-zero spin are expected to order magnetically sufficiently close to absolute zero (– 273.15 °C). The critical temperature T_c, below which the substance must be cooled before spontaneous order occurs, depends on the strength of the spin-spin interactions. Since the nuclear magneton is small, the required temperatures are in the sub-microkelvin range. There are a few exceptions of spontaneous nuclear ordering in the millikelvin region, most notably solid ^{3}He (1) and Van Vleck paramagnets (2) in which T_c is increased by zero point fluctuations and by hyperfine enhanced internal fields, respectively.

In 1951, Purcell and Pound first produced negative temperatures, using LiF as the working substance (3). The implications of these early NMR experiments have been discussed by Ramsey and by Van Vleck (4). Beginning in 1968, nuclear co-operative phenomena at positive and negative temperatures have been investigated at Saclay (France), under the leadership of Abragam and Goldman, in dielectric materials like CaF_2 and LiH (5). Magnetic ordering in $Ca(OH)_2$ has been studied by Wenckebach and his coworkers (6).

Experiments on nuclear magnetic ordering in metals are based on the pioneering studies of Kurti and his coworkers (7). They established, as early as 1956, the feasibility of the nuclear demagnetization method. In spite of the limitations imposed by cryogenic techniques available at that time, the Oxford group succeeded in reaching 1 μK in the nuclear spin system of copper. Subsequent improvements in experimental procedures, after the advent of powerful dilution refrigerators and superconducting magnets in the late sixties, have made nuclear cooling a reality even below 1 nK.

Spontaneous nuclear magnetic ordering has been studied extensively at $T > 0$ in copper and more recently at $T > 0$ and at $T < 0$ in silver and rhodium (8). This type of experiments at ultralow temperatures have been carried out at the Hel-

sinki University of Technology for almost twenty years (9). In copper, competition between the dipolar force and the conduction-electron-mediated exchange interaction leads at $T > 0$ to three different antiferromagnetic structures. At negative spin temperatures, ferromagnetic order has been observed in silver (10). In this metal, the phase transitions occur at 560 pK (1 pK = 10^{-12} K) and at –1.9 nK, respectively. In rhodium, spin temperatures of 280 pK and – 750 pK have been reached (11); these are the lowest and 'highest' temperatures ever produced and measured.

Nuclear Magnetism in Metals

The Hamiltonian of nuclei in silver metal can be written in the form $\mathcal{H} = \mathcal{H}_{dip} + \mathcal{H}_{ex} + \mathcal{H}_Z$. The dominating spin-spin energy is the nearest-neighbor antiferromagnetic exchange interaction $\mathcal{H}_{ex}$ (12). The dipolar force $\mathcal{H}_{dip}$ is smaller by a factor of three and the Zeeman term $\mathcal{H}_Z$ is proportional to the external magnetic field B. Owing to the strong exchange interaction, the spin system in silver bears a close resemblance to an fcc Heisenberg antiferromagnet which has been the object of much theoretical interest; the ground state properties are affected by 'frustration' (13). Because the nuclear spin $I = {}^1/_2$, quantum effects are expected to be prominent.

In rhodium the *d*-electron-mediated anisotropic exchange forces, characteristic of transition metals (14), contribute as well. These can be taken into account, approximately, by adding a pseudodipolar term $\mathcal{H}_{psd}$ to the Hamiltonian. The presence of close competition between isotropic and anisotropic spin-spin interactions permits a broad spectrum of magnetic configurations in Rh. A somewhat similar situation exists in copper, where dipolar and exchange interactions compete. Neutron diffraction experiments on Cu (15) have revealed up-up-down spin modulations whose magnitude depends on the external magnetic field. In-

teresting results of nuclear ordering have been obtained on scandium metal and on $AuIn_2$ (16). The investigations of nuclear spin systems may thus be compared with studies of rare earth metals which have revealed many novel spin configurations, even during recent years (17).

All experiments on insulators (5, 6), performed using dynamic nuclear polarization followed by adiabatic demagnetization in the rotating frame, are limited to ordering by the truncated dipolar force. The main weakness of the dynamic method of cooling is, however, the inevitable presence of electronic paramagnetic impurities, introduced purposely for nuclear polarization by means of the 'solid effect'; the strong local fields produced by the impurities probably blur, to a certain extent, some of the features of the nuclear long-range order. Silver and rhodium provide more general systems for experimental and theoretical studies of nuclear magnetism at $T > 0$ and at $T < 0$.

Negative Absolute Temperatures

Ordinarily, the temperature in kelvins describes the average energy which is either connected with the free motion of the particles or with their vibration about the lattice sites. In both cases the energy per particle is on the order of $k_B T$. The kinetic energy has no upper bound. Thus, if the temperature were raised towards infinity, the energy of the system would increase without limit. This means that the temperatures for electrons and for the lattice are limited to $0 < T < \infty$ since all the possible values of energy are covered by this range.

Absolute temperature determines the distribution of particles on the available energy levels according to the Boltzmann factor, $\exp(-E/k_B T)$. In systems, where the energy levels are bound from above, it is possible to have negative temperatures because the Boltzmann factor does not diverge in this case. A

negative temperature describes a state with population inversion where the higher energy levels have more particles than the lower levels.

The theorems and procedures of statistical mechanics, such as the use of the partition function and the quantum mechanical density matrix, apply equally to systems at negative temperatures. By examining the statistical theory by which the Boltzmann distribution is derived, there is nothing objectionable *a priori* for the parameter $1/k_B T$ being negative; $T < 0$ simply means that the mean energy of the system is higher, instead of being lower than that corresponding to equal populations among the energy levels at $T = \pm\infty$.

From Minimum to Maximum

The energy level diagram for an assembly of silver or rhodium nuclei is illustrated schematically in Fig. 1; the spin $I = {}^1/_2$, so there are just two levels, corresponding to the nuclear magnetic moment $\boldsymbol{\mu}$ parallel and antiparallel to the external field $\boldsymbol{B}$, respectively. The distribution of the nuclei among the Zeeman energy levels is again determined by the Boltzmann factor, $\exp(\boldsymbol{\mu}\cdot\boldsymbol{B}/k_B T)$, using $E = -\boldsymbol{\mu}\cdot\boldsymbol{B}$. At positive temperatures the number of nuclei in the upper level, with $\boldsymbol{\mu}$ antiparallel to $\boldsymbol{B}$, is always smaller than in the lower. At the absolute zero, all nuclei are in the ground level with $\boldsymbol{\mu}$ parallel to $\boldsymbol{B}$.

As the temperature is increased from $T = +0$ (lower row from left to right in Fig. 1), nuclei flip into the upper energy level and, at $T = +\infty$, there is an equal number of spins in both levels; the infinite temperature, however, does not cause any problems since the level spectrum has an upper limit. When the energy is increased further (upper row from right to left) by lifting more spins to the higher level, the inverse spin distribution can still be described by the Boltzmann factor but now with $T < 0$. Finally, when approaching zero from the nega-

tive side, $T \rightarrow -0$, eventually only the upper energy level is populated. Since heat is transferred from the warmer to the colder body when two systems are brought into thermal contact, negative temperatures are actually 'hotter' than positive ones.

Near the absolute zero, $1/T$ or $\log T$ is sometimes used as the temperature function but, when $T < 0$, $\log T$ is not suitable (4). However, on the inverse-negative scale, $\beta = -1/T$, the coldest temperature, $T = +0$, corresponds to $\beta = -\infty$ and the hottest temperature, $T = -0$, to $\beta = +\infty$. On this scale the algebraic order of β and the order from cold to hot are identical; the system passes from positive to negative temperatures through $\beta = -0 \rightarrow +0$. This inverse-negative scale thus runs in an 'orderly' fashion from the coldest to the hottest. The third law of thermodynamics emerges 'naturally' by the impossibility to reach the positive or negative end of the β-axis.

At $T = +0$, an isolated nuclear spin assembly has the lowest and, at $T = -0$, the highest possible energy (see Fig. 1). This important fact can be put into a more general basis. During demagnetization, the external magnetic field $\boldsymbol{B}$ at first completely controls the order in the nuclear spin system. However, when the field has been reduced sufficiently, approaching the internal local field B_{loc} and ultimately even $B = 0$, the dipole-dipole and exchange forces gradually take over and the spin order begins to change from that forced by $\boldsymbol{B}$ to an arrangement determined by mutual interactions. During this spontaneous adjustment of spins the entropy increases, according to the general principles governing thermodynamic equilibrium, until S reaches a maximum while the magnetic enthalpy $H = U - BM$ stays constant.

In order to find the equilibrium spin configuration, one has to consider the variation of entropy under the restriction of a constant enthalpy, that is, one must seek an extremal value of $S + \beta H$ where, by differentiation, Lagrange's multi-

plier $\beta = -\,dS/dH = -1/T$. Thus, one obtains $S - H/T = -\,G/T$ for the thermodynamic potential reaching an extremum; G is the Gibbs free energy. When $T > 0$, $G = H - TS$ and the extremum is a minimum since S assumes its maximal value at equilibrium. When $T < 0$, $G = H + |T|S$ and the Gibbs free energy obviously reaches a maximum.

The tendency to maximize energy, instead of minimizing it, is the basic difference between negative and positive temperatures. In silver, the nearest-neighbor antiferromagnetic Ruderman-Kittel exchange interaction (18), three times stronger than the dipolar force, favors antiparallel alignment of the nuclear magnetic moments and thus leads to antiferromagnetism when $T > 0$. At $T < 0$, since the Gibbs free energy now must be maximized, the very same interactions produce ferromagnetic nuclear order. The long-range dipolar force favors the formation of ferromagnetic domains. In rhodium the situation is more complicated (see below).

Experimental Methods

Two different temperatures are simultaneously applicable in solids at ultra low temperatures. In metals there is only a loose thermal contact between conduction electrons and the nuclei; for example, in silver and rhodium 14 hours are needed to reach equilibrium between the two systems at an electronic temperature $T_e = 200\,\mu K$. Since the spin-spin relaxation time $\tau_2 = 10$ ms, the nuclei can quickly equilibrate among themselves to a common spin temperature. Therefore, on time scales 10 ms $<<$ t $<<$ 20 h, two separate temperatures exist in the metal: T_e for the lattice and conduction electrons and T for the nuclei. In silver and rhodium metals, the spin-lattice relaxation time $\tau_1 = (10\ \mathrm{sK})\,/\,T_e$. Thus, for good thermal isolation between conduction electrons and the nuclei, a low T_e is

needed; this is why the experiments must be carried out at ultra low temperatures.

In dielectric materials it is rather easy to generate negative temperatures. By employing the 'solid effect' (19), inverted spin polarizations can be produced, with the help of electronic magnetic impurities, by rf-irradiation at $\nu_e + \nu_n$, where ν_e and ν_n are the electronic and nuclear Larmor frequencies, respectively. Initially (5), at $T_i = 0.7$ K and $B_i = 2.7$ T, the polarization of nuclear moments is almost zero whereas the more than 1000-times larger electronic moments are polarized close to 100%. The rf-field induces simultaneous 'flip-flip' transitions of electronic and nuclear moments to higher energy levels, where they are aligned antiparallel to $\boldsymbol{B}$. An inverted electronic spin 'flops' quickly back to its original state and is immediately ready to flip another nucleus. Since coupling between the nuclei and the lattice is weak at the low operating temperature and high magnetic field, nuclear relaxation is slow.

Spin polarizations up to 90% ($p = -0.90$) have been obtained by this method in CaF_2 (5). Nuclear demagnetization must then be carried out in the rotating coordinate frame, which selectively keeps the electronic spins in a high field and thereby prohibits energy exchange between the nuclear and electronic systems, that is, prevents loss of nuclear polarization. The Saclay group of Abragam and Goldman has produced negative spin temperatures, probably 'above' -1 μK, but the temperatures were not directly measured. Spontaneous nuclear order, both at $T > 0$ and at $T < 0$, was observed (5).

Population inversion from $T > 0$ to $T < 0$ is much harder to generate in metallic samples for two reasons: Substantial effort is needed to reach the high initial spin polarizations and eddy currents make the production of inverted spin populations a difficult task. In spite of these problems, Oja, Annila, and Takano in our laboratory, decided to try an experiment on silver (20): The external mag-

netic field $\boldsymbol{B}$ was reversed quickly, in a time $t \leq \tau_2 = 10$ ms, so that the nuclei would not have a chance to rearrange themselves among the energy levels. This simple idea worked: Negative spin temperatures were produced, but the loss of polarization was large. After gradual improvements and refinements in the technique, by Hakonen and his coworkers, fully satisfactory results were obtained, first on silver (10) and later on rhodium (11).

Indeed, it is important to realize that the field flip must be rapid in comparison to τ_2, the Larmor period of the spins in the local field B_{loc}. If this condition is not met, the spins are able to follow adiabatically the field reversal, and negative temperatures will not result. Demagnetization would just be followed by remagnetization to the positive starting temperature. In fact, during the quick field flip the Boltzmann distribution of the spins breaks down and, for a short moment, the system cannot be assigned a temperature. In a certain sense, the spin assembly passes from positive to negative temperatures via $T = +\infty = -\infty$, without crossing the absolute zero. Therefore, the third law of thermodynamics is not violated.

To obtain nuclear temperatures in the nano- and picokelvin regime, a sophisticated 'brute force' cooling apparatus, with two nuclear refrigeration stages in cascade, was employed in Helsinki (8). The second stage was the sample itself. The cooling procedure is illustrated schematically and explained in some detail by Fig. 2. The experiment must be carried out quickly after the final demagnetization, since the nuclear spin temperature starts immediately to relax towards T_{e} with the time constant τ_1, determined by the spin-lattice relaxation process.

One of the difficult problems in these experiments was to measure the absolute temperature of the nuclei. The second law of thermodynamics, $T = \Delta Q / \Delta S$, was used directly. When $T > 0$, the nuclear spin system was supplied with a small amount of heat ΔQ and the ensuing entropy increase ΔS was calculated from the

measured loss of nuclear polarization p. At negative temperatures, $\Delta Q < 0$ when entropy increases: the system radiates energy at the nuclear Larmor frequency while the populations of the two energy levels tend to equalize.

NMR Emission at $T < 0$

The quantity that was actually measured in the Helsinki experiments (20, 21) is the NMR absorption χ'' recorded by a SQUID. Nuclear polarization was computed from $p = A\int\chi''(\nu)\mathrm{d}\nu$, where ν is the NMR excitation frequency and the constant A was determined from measurements near 1 mK. The NMR data (see Fig. 3) showed that instead of absorption, as at $T > 0$, the system was emitting energy when $T < 0$.

In Fig. 4, the absolute value of the inverse magnetic susceptibility $|1/\chi|$ of silver, calculated from the Kramers-Krönig relation $\chi = (1/\pi)\int(\chi''/\nu)\mathrm{d}\nu$, is plotted as a function of $|T|$ in nanokelvins (21). We note that at positive temperatures one obtains a straight line with an intercept on the negative side of the $|T|$-axis. This behavior is typical and indicates that silver tends to antiferromagnetic order when $T \rightarrow +0$. In later experiments (22), proof of a transition at $T_N = 560$ pK came from changes in the NMR spectra upon ordering.

At negative temperatures, the intercept is on the positive side of the $|T|$-axis, which shows that, when $T \rightarrow -0$, the spin system of silver nuclei tends to ferromagnetic order, as expected; the transition point, however, was not reached in these early experiments. The data in Fig. 4, both at $T > 0$ and at $T < 0$, follow the Curie-Weiss law down to the lowest temperatures. This rather uncommon feature has been reproduced theoretically, starting from first-principles band structure calculations (23).

Later, the spontaneously ordered phase was reached at $T < 0$ (10). The static magnetic susceptibility of silver spins was found to saturate at $\chi_{sat} = -1.05$ (see Fig. 5), which is a typical value for ferromagnetic ordering into a domain state, caused by dipolar interactions. Within the scatter of experimental data, the critical polarization $p_c = -0.49$ was constant below 5 μT, for magnetic fields both parallel and perpendicular to the sample foils. By employing the linear relationship, $1/|p| - 1 = 0.55(|T|/\text{nK})$, between the inverse polarization and temperature, the Curie point $T_C = -1.9 \pm 0.4$ nK was obtained.

Phase Diagram of Silver

The magnetic field versus entropy diagram of silver, for $T > 0$ and $T < 0$, is shown in Fig. 6 (10, 22). The difference at positive and negative temperatures reflects frustration of antiferromagnetic interactions (13) as well as the influence of dipolar forces which favor ferromagnetism. The critical field B_c of the ferromagnetic phase is determined by the strength of dipolar forces, while B_c of the antiferromagnetic state is caused by the magnitude of the exchange energy. The bulge in the phase diagram at $T > 0$, indicating that a small field favors antiferromagnetic ordering in silver, does not have a satisfactory explanation at present.

The saturation of susceptibility to -1 in the ordered state can be explained only by the formation of domains, since otherwise χ_{sat} would diverge at T_C. Instead of needles, as at $T > 0$, plate-like domains are expected when energy is maximized at $T < 0$. A comparison of the saturation values of susceptibility with mean-field and Monte-Carlo calculations by Viertiö and Oja (24) indicates that in fields parallel to the sample foils there is a multidomain structure in silver, while in transverse fields a single-domain configuration is preferred.

The domain structure at $T < 0$ is illustrated in the insert of Fig. 6. The direction of magnetization $\boldsymbol{M}$ is degenerate, but the orientation of $\boldsymbol{M}$ in one of the domains determines the whole spin arrangement, because the tangential component of $\boldsymbol{M}$ has to be continuous and the perpendicular component must change sign across a domain wall. Moreover, the total magnetization has to satisfy the condition $\chi_{\mathrm{sat}} \approx -1$.

Rhodium Prefers Antiferromagnetism

The absolute value of the inverse static susceptibility of Rh nuclei as a function of $|T|$ is shown in Fig. 7 (11). The solid line represents the antiferromagnetic Curie-Weiss law, $\chi = C/(T - \theta)$, with $C = 1.3$ nK and $\theta = -1.4$ nK, obtained at positive temperatures from the low polarization data. At $T < 0$, the corresponding ferromagnetic dependence is displayed by the dashed line. At low temperatures the Curie-Weiss approximation is known to deviate, especially when $I = {}^1/_2$, from the more accurate results based on high-T series expansions (25). For negative temperatures, the measured data (●) show a crossover from ferro- to antiferromagnetic behavior around -5 nK. This indicates that the energy of nuclear spins in rhodium is both minimized and maximized by antiferromagnetic order.

The data on Rh at $T > 0$ and at $T < 0$ extend to roughly a factor of two closer to the absolute zero than the temperatures reached in the experiments on silver. Phase transitions, however, were not seen in rhodium, even though the experimentally produced polarizations of Rh nuclei, $p = 0.83$ and $p = -0.60$ at $T > 0$ and at $T < 0$, respectively, were higher than those required for ordering in silver. This is an indication that in rhodium nearest and next-nearest neighbor interactions are of almost equal magnitude but of opposite sign. The transition temperature is thus very low, which explains why no ordering was detected in spite

of the extreme positive (280 pK) and negative (–750 pK) spin temperatures produced in rhodium.

The susceptibility data can be used to extract the nearest and next-nearest neighbor Heisenberg interaction coefficients J_1 and J_2 (11); the values obtained from experimental results are $J_1/h = -17$ Hz and $J_2/h = 10$ Hz. Molecular-field calculations have been employed to predict regions of different types of magnetic ordering in the J_2 *versus* J_1 -plane (26). In an fcc lattice, ferromagnetism is present only when $J_1 > 0$ and $J_2 > -J_1$. The antiferromagnetic part is divided to Type-I, Type-III, and Type-II regions at $J_2 = 0$ and at $J_2 = J_1/2$, so that rhodium lies well inside the Type-I region at $T > 0$ (see Fig. 8). At $T < 0$, the signs of the J's are effectively reversed, and the corresponding point (●) is located in the ferromagnetic sector, rather close to the Type-II antiferromagnetic border. Better data on the interaction parameters are needed to resolve the discrepancy with Fig. 6, especially since improved theoretical calculations (27) have recently become available.

Spin-lattice relaxation times, measured at positive and negative temperatures, have been investigated in rhodium where iron impurities shorten τ_1 substantially in small magnetic fields (28). These results have revealed an unexpected asymmetry in τ_1 with respect to the sign of the nuclear spin temperature: The spin-lattice relaxation has a contribution which varies as $1/T$, but τ_1 is longer when $T < 0$. This finding is hard to interpret since all theories (29) predict symmetric behavior. The experimental result might be a screening effect caused by the large negative susceptibility at $T < 0$, in a similar way as screening is produced by the Meissner effect in superconductors.

Concluding Remarks

By means of NMR measurements it is not possible to verify the details of magnetically ordered structures. Experiments employing neutron scattering are necessary for this purpose. A joint effort between the Hahn-Meitner-Institut in Berlin, our Laboratory, the Risø National Laboratory (Denmark), and the University of Copenhagen has been started in order to perform neutron diffraction experiments on magnetically ordered silver at $T > 0$ and perhaps at $T < 0$ as well. These measurements are being carried out in the group of Michael Steiner in Berlin.

In copper τ_2 is 100 times shorter than in silver and rhodium. Therefore, producing spontaneous nuclear order at $T < 0$ has not succeeded in this metal, because the external field could not be flipped fast enough without causing massive eddy current heating in the specimen. However, we believe that besides silver and rhodium, it is possible to reach magnetic ordering at negative spin temperatures in other metallic systems which have spin-spin relaxation times on the order of 1 ms.

This provides interesting experimental possibilities. For example, the interplay between superconductivity and magnetism could be investigated: By reversing the sign of temperature, the nuclear spin order might be changed from antiferromagnetism to ferromagnetism or vice versa, and the effect of this transformation on the superconducting properties could be investigated. Unfortunately, owing to supercooling, experiments of this type did not succeed in Rh, even though T_e in the Helsinki experiments (11) was considerably lower than 325 μK, the critical temperature for superconductivity in rhodium (30).

It has been argued, sometimes, that negative temperatures are fictitious quantities because they do not represent true thermal equilibrium in a sample consist-

ing of nuclei, conduction electrons, and the lattice. However, the experiments on silver, in particular, show conclusively that this is not the case. The same interactions produce ferro- or antiferromagnetic order, depending on whether $T < 0$ or $T > 0$. Besides, true equilibrium, in the strictest sense of the word, hardly ever exists in nature.

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31. We thank A.S. Oja for comments on the manuscript. This work was sponsored by the Academy of Finland.

Figure Captions

Fig. 1. Energy level diagram of silver (or rhodium) nuclei in a constant magnetic field for $T > 0$ (lower row) and $T < 0$.

Fig. 2. Schematic illustration, on a temperature *versus* entropy diagram, of the procedure for cooling an assembly of silver or rhodium nuclei to negative nanokelvin temperatures.

(A → B) Both nuclear stages are cooled to T_i = 15 mK by the dilution refrigerator and, simultaneously, the first stage is polarized in a strong magnetic field B_i = 8 T.

(B → C) The nuclei of the first stage, made of 20 mol of copper, are adiabatically demagnetized to B_f = 100 mT, which produces a low temperature $T_f = T_i(B_f/B_i) \approx 200$ μK. Towards the end of demagnetization, the second nuclear stage, that is, the sample, is magnetized to 8 T.

(B → D) The 2-gram silver or rhodium specimen of thin polycrystalline foils cools in the field B = 8 T, by thermal conduction, to $T_f \approx 200$ μK.

(D → E) The specimen is demagnetized from 8 T to 400 μT, whereby the spins cool into the low nanokelvin range ($T \approx (400\,\mu\text{T}/8\,\text{T})\,200\,\mu\text{K} = 10$ nK), thermally isolated by the slow spin-lattice relaxation (τ_1 = 14 h) from the conduction electrons which are anchored to 200 μK by the first nuclear stage at C. By continuing demagnetization of the specimen to zero field (not shown in the diagram), the record temperature of 280 pK was reached in rhodium. In silver, dipole-dipole and exchange interactions produced antiferromagnetic order at the Néel temperature T_N = 560 pK.

(E → F) Finally, the negative spin temperature is achieved in the system of silver or rhodium nuclei by reversing the 400-μT magnetic field in about 1 ms. The rapid inversion causes some loss of polarization (that is, increase of entropy). By continuing demagnetization to zero field, the record temperature of

– 750 pK was reached in rhodium (not shown in the diagram). In silver, dipole-dipole and exchange interactions produce ferromagnetic order at the Curie temperature $T_C = -1.9$ nK.

(F → G → A) The system starts to lose its negative polarization, crossing in a few hours, via infinity, from negative to positive temperatures.

(C → A) The first nuclear stage warms slowly, under the $B_f = 100$ mT field, from $T_f = 200$ μK towards 15 mK. A new experimental sequence can then be started.

Fig. 3. NMR absorption (blue) and emission (red) spectra of silver nuclei measured at $T = 1$ nK and $T = -4.3$ nK, respectively (21). The imaginary component of susceptibility χ'' has been plotted against the NMR frequency ν.

Fig. 4. Absolute value of the inverse static susceptibility versus absolute value of the spin temperature for nuclei in silver at $T > 0$ (○) and at $T < 0$ (●) (21).

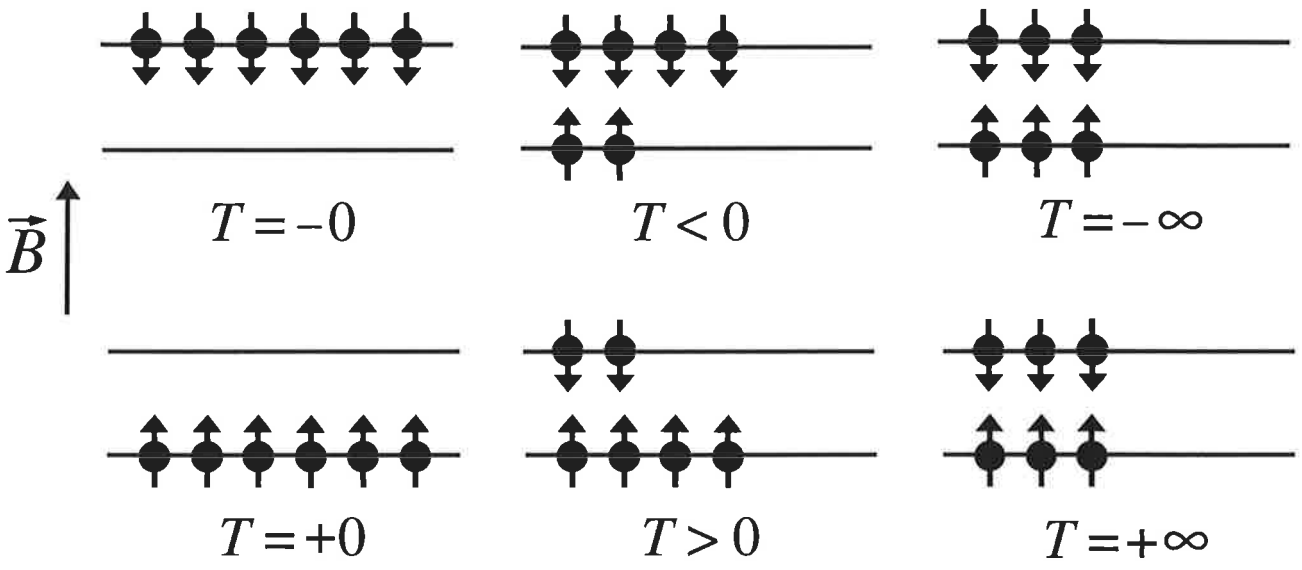
Fig. 5. Static susceptibility of silver as a function of polarization p of nuclear spins at $T < 0$, measured in zero field (○) and at $B = 5$ μT oriented perpendicular (▽) and parallel (△) to the sample foils (10). The fitted curve represents the Curie-Weiss law, and the straight horizontal line corresponds to the saturation value of χ in the ordered state as predicted by mean-field calculations (24). The scale on top gives estimated temperatures which apply only at $B = 0$.

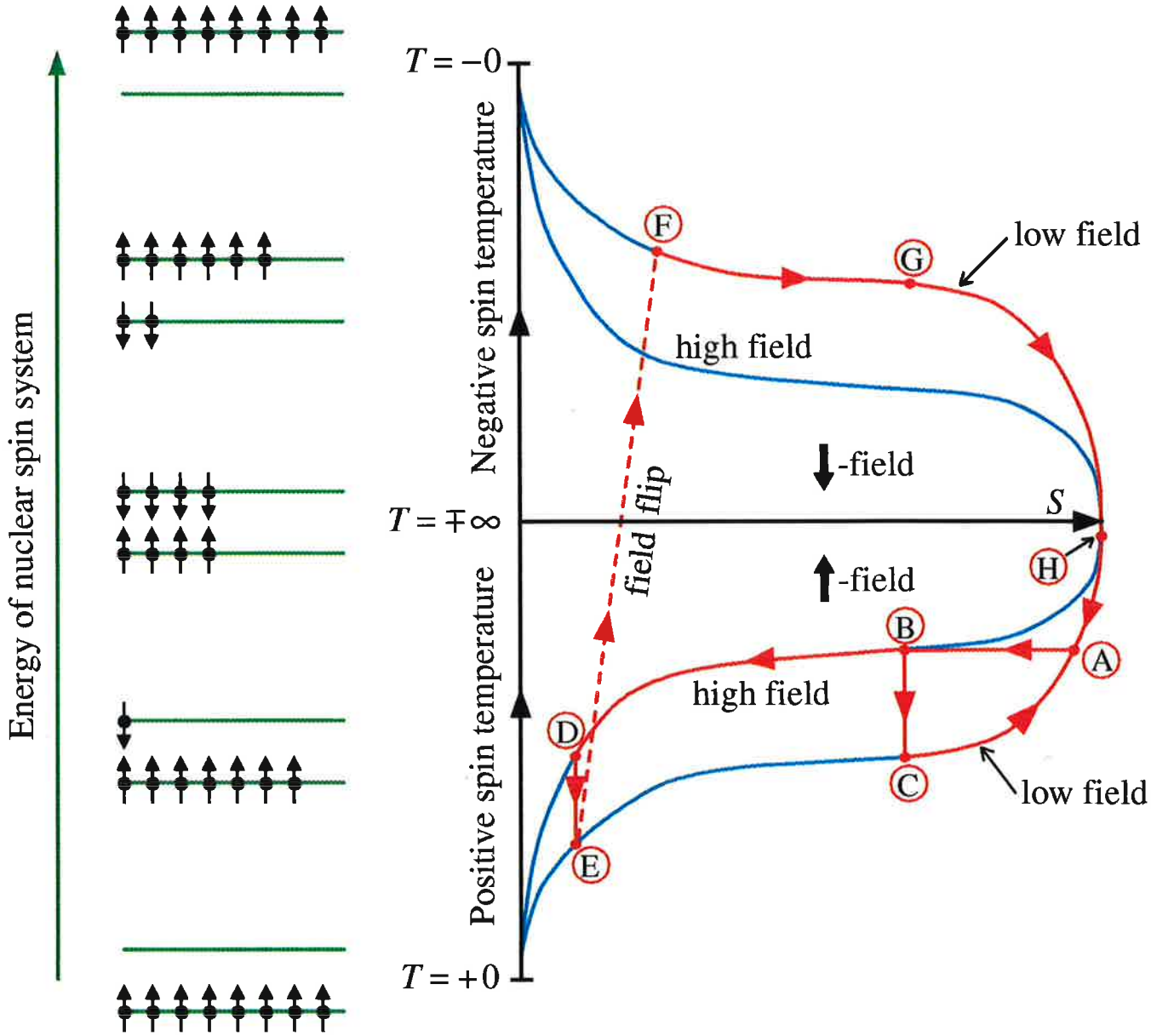
Fig. 6. Phase diagram of nuclear spins in silver metal for negative (red) and positive (blue) absolute temperatures in the magnetic field *versus* reduced entropy ($S/\mathcal{R}\ell$n2) plane (10). The ferromagnetic spin arrangement at $T < 0$ is illustrated in the insert. The size of the domains is large compared with the interatomic spacing but small with respect to the dimensions of the sample.

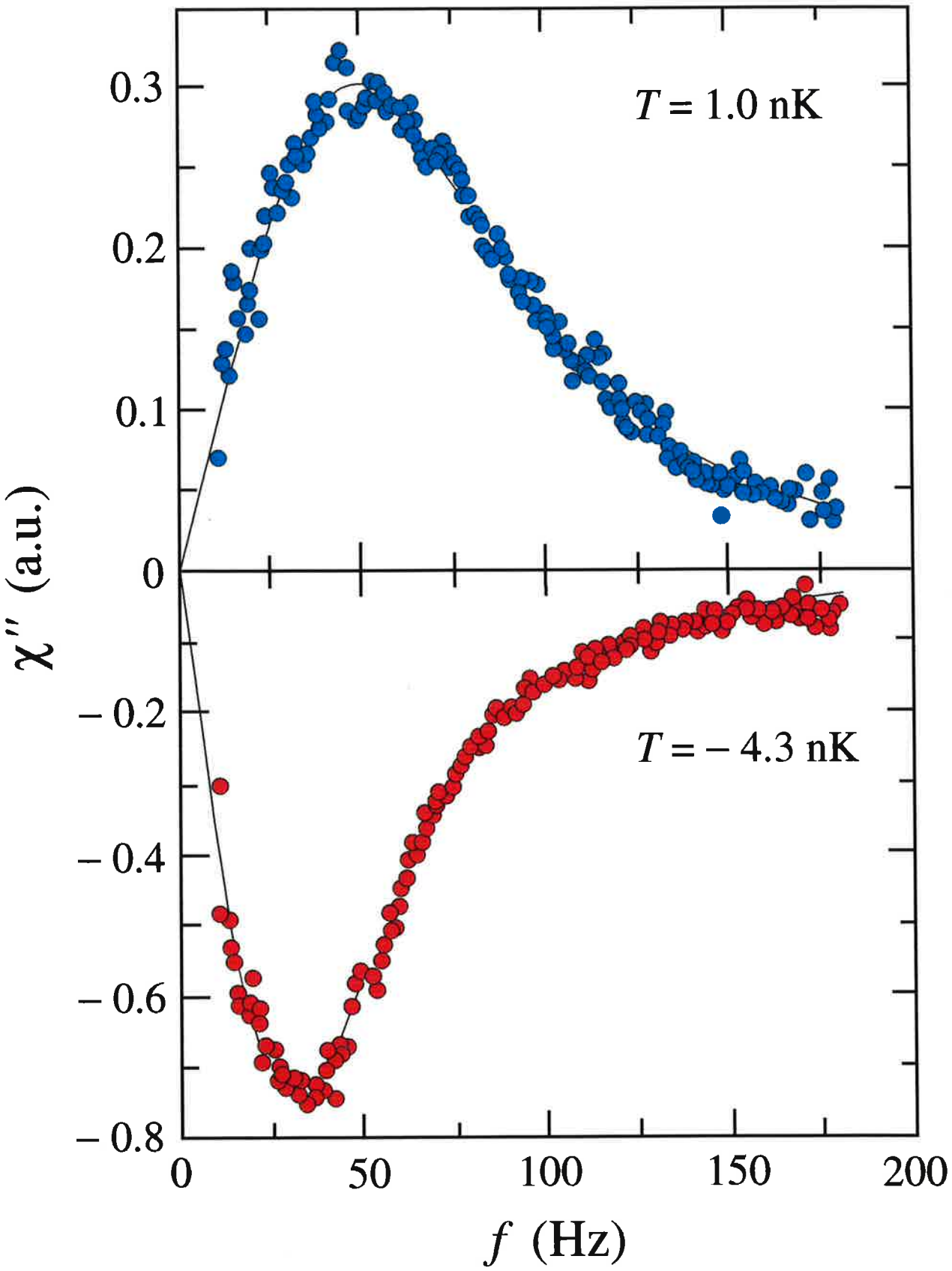
Fig. 7. Absolute value of the inverse static susceptibility versus absolute value of the spin temperature for nuclei in rhodium at $T > 0$ (○) and at $T < 0$ (●) (11). The external magnetic field $B = 0$.

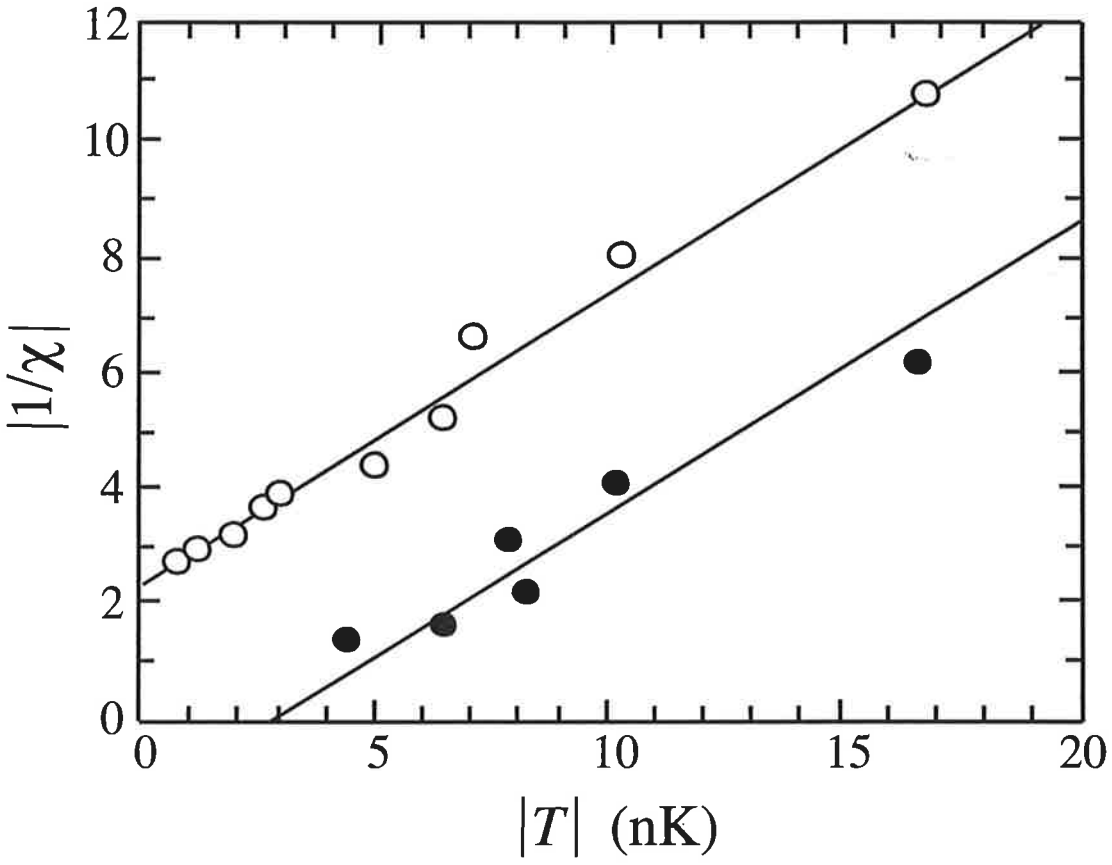
Fig. 8. Phase diagram of magnetic ordering in the J_2 *versus* J_1 -plane as predicted by the mean field theory for an fcc-lattice (26). Antiferromagnetic regions of Type-I, -II, and -III are denoted by AF1, AF2, and AF3, respectively, whereas FM refers to ferromagnetic ordering. Interaction parameters for Rh (○) and Ag (Δ) are plotted in the figure; open and filled symbols refer to positive and negative temperatures, respectively. The different antiferromagnetic spin structures are described in Ref. 26.

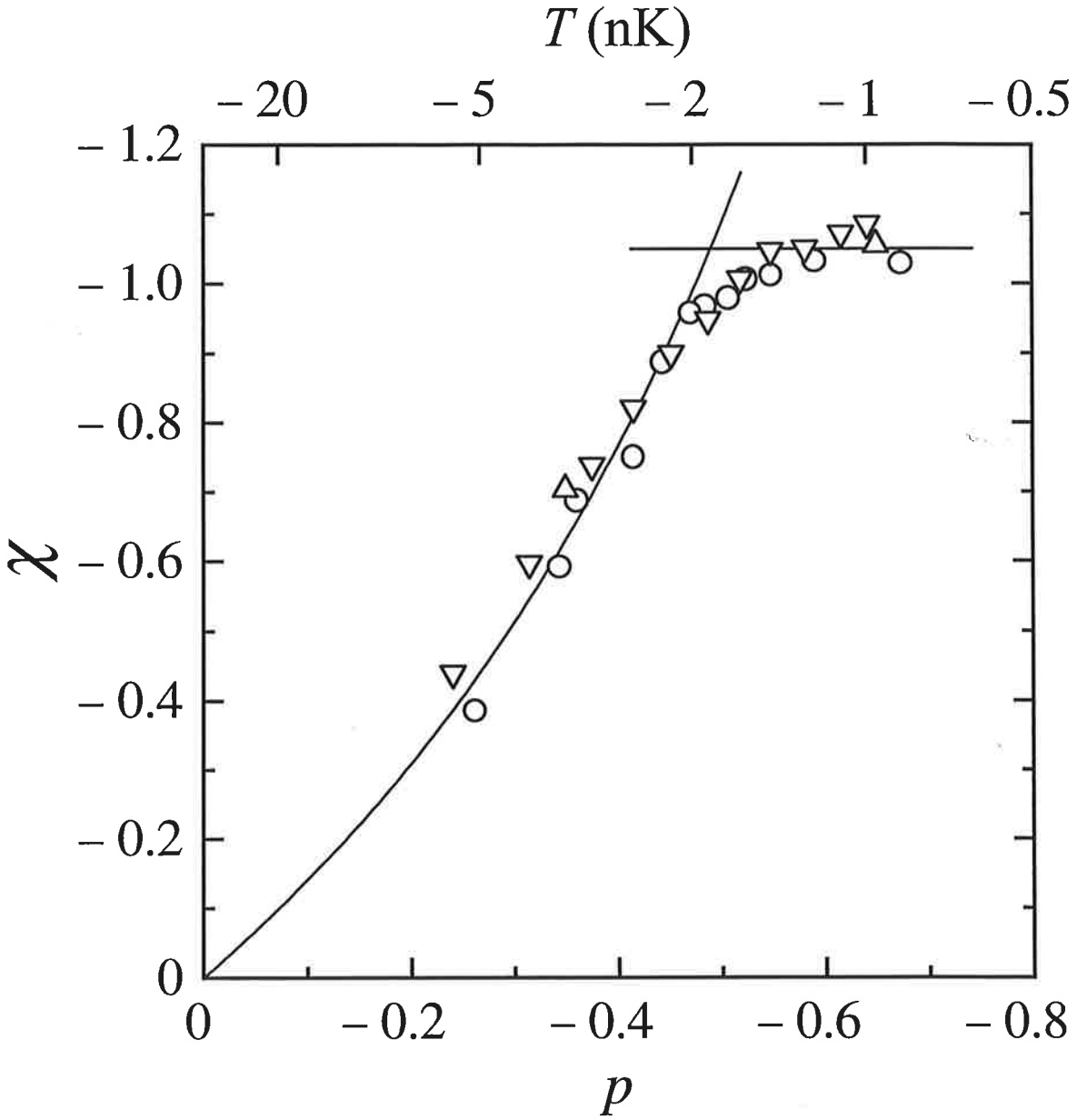
Total number of words 5583

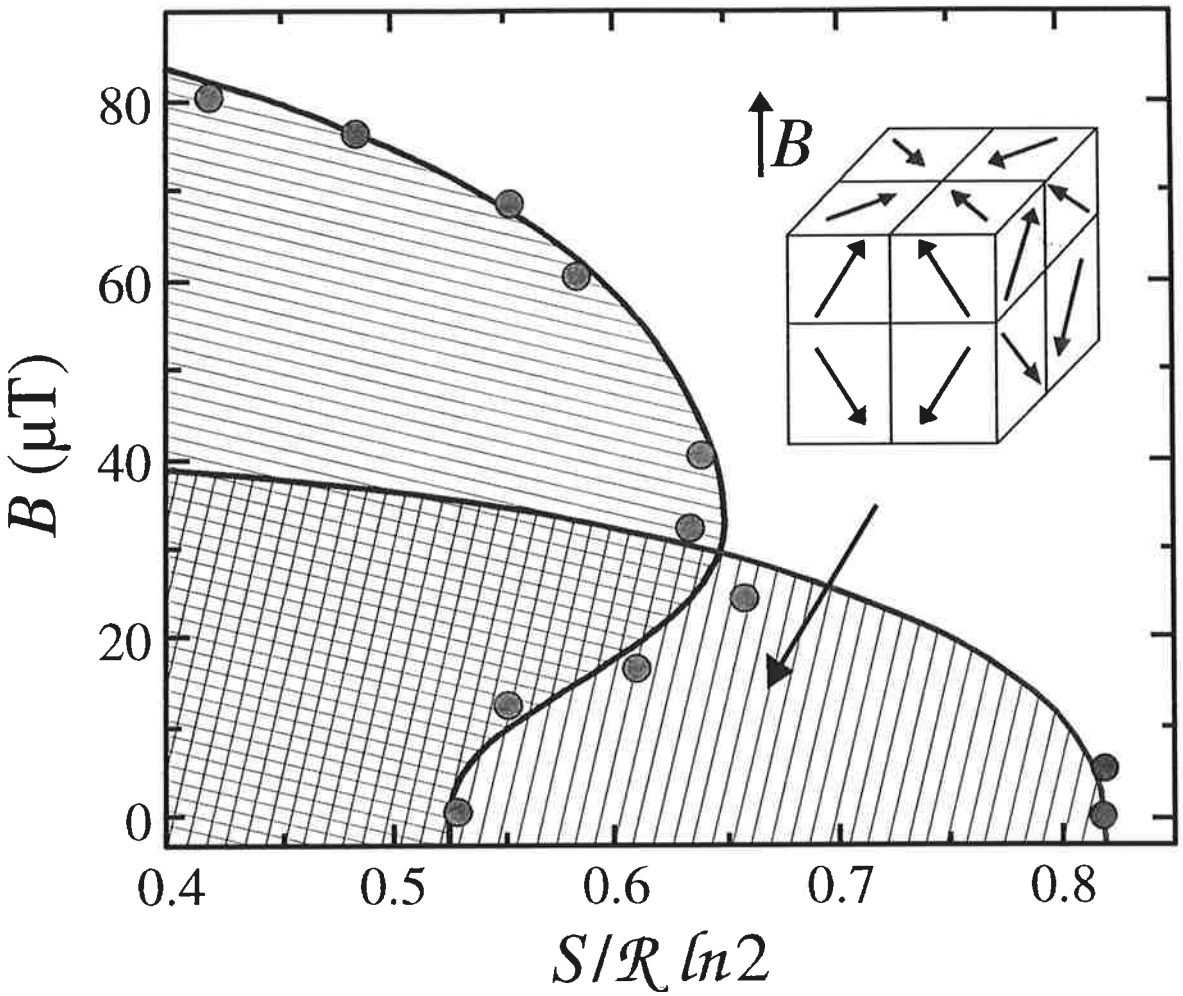

$\vec{B}$
$T=-0$
$T<0$
$T=-\infty$
$T=+0$
$T>0$
$T=+\infty$

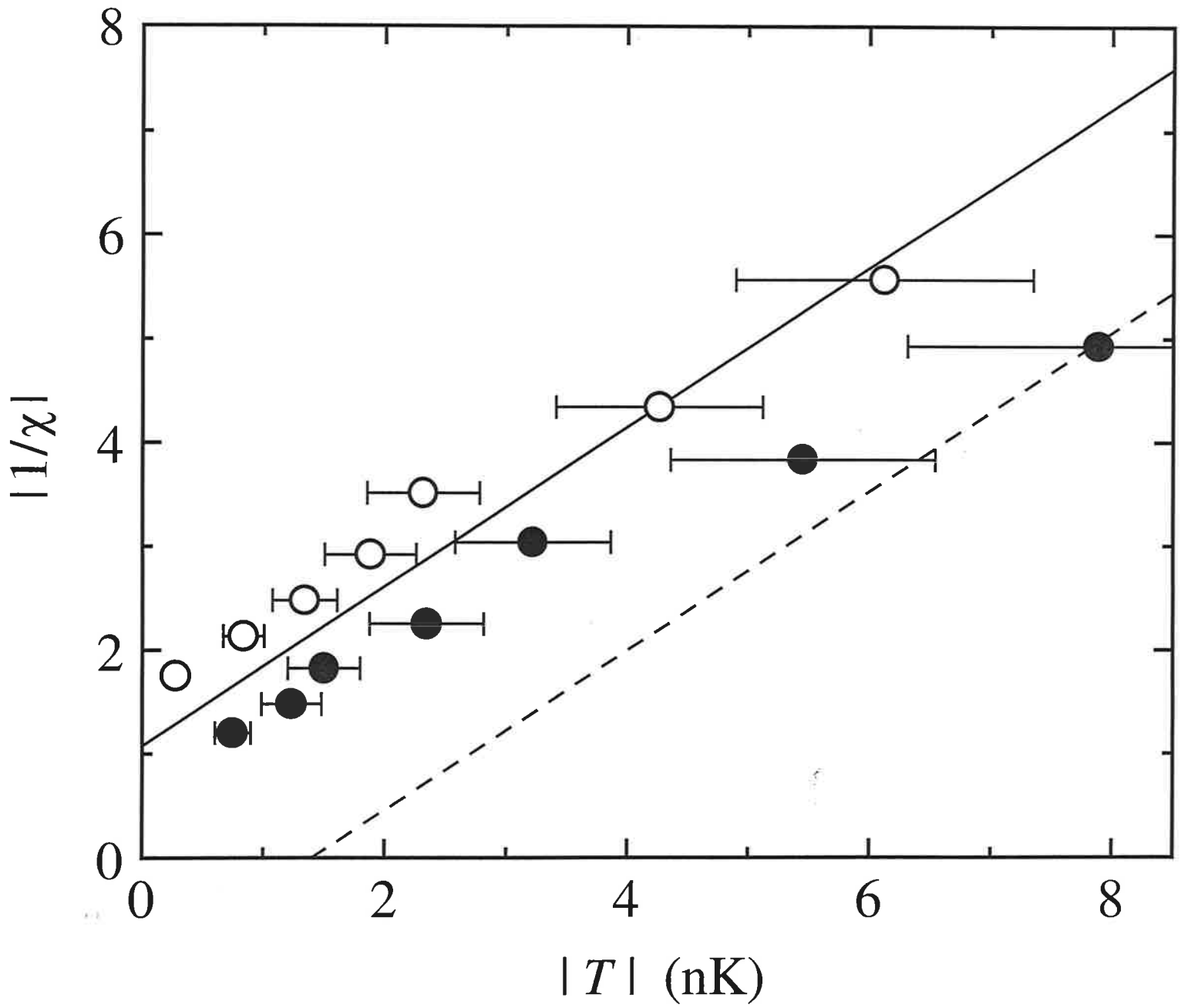

Energy of nuclear spin system
T = −0
Negative spin temperature
T = ∓∞
Positive spin temperature
T = +0
F
G
low field
high field
-field
S
field flip
H
-field
B
A
high field
D
C
low field
E

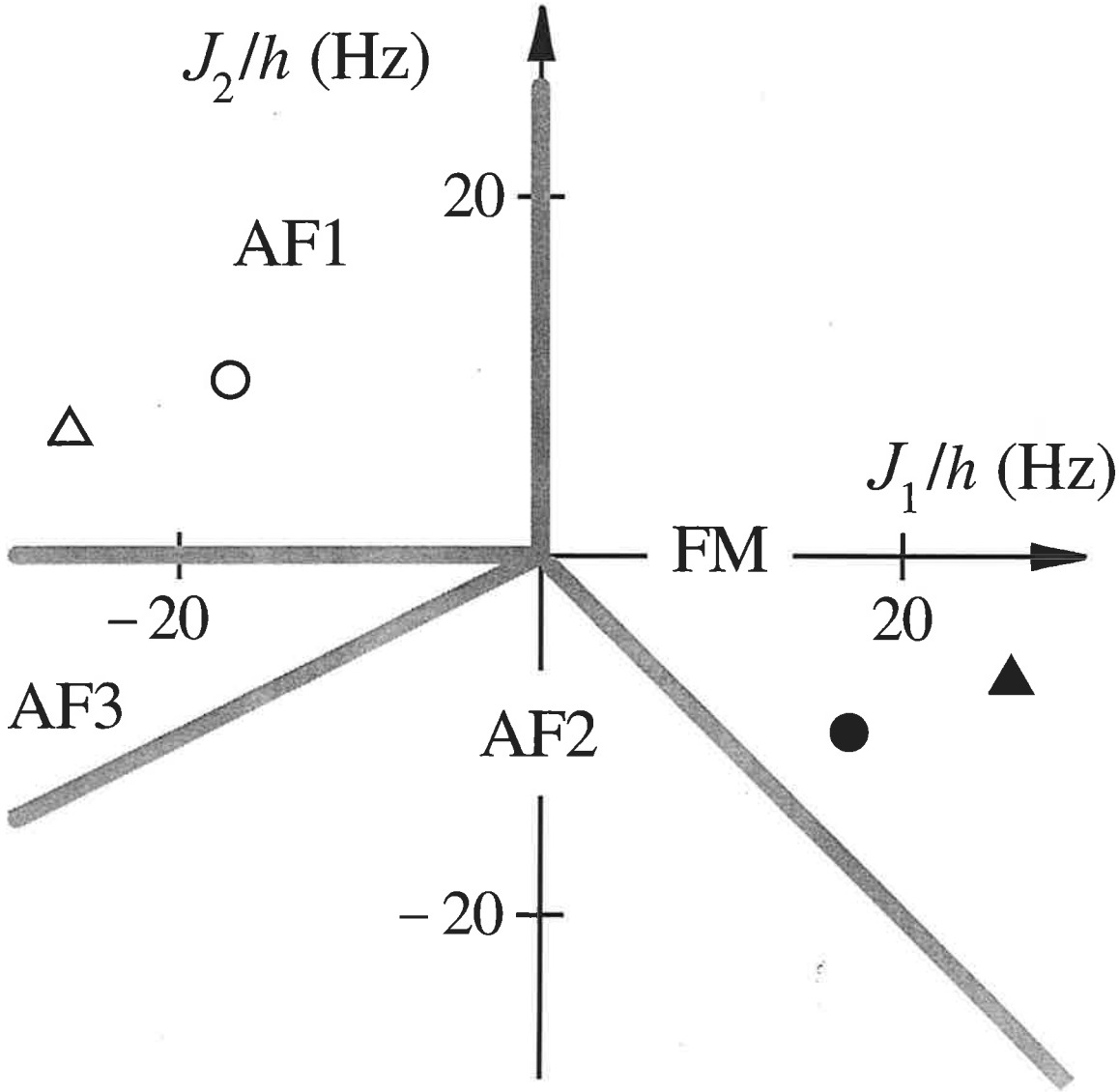

0.3
0.2
0.1
0
– 0.2
– 0.4
– 0.6
– 0.8
χ'' (a.u.)
T = 1.0 nK
T = – 4.3 nK
0
50
100
150
200
f (Hz)


12
10
8
6
4
2
0
0
5
10
15
20
|1/χ|
|T| (nK)


T (nK)
– 20
– 5
– 2
– 1
– 0.5
– 1.2
– 1.0
– 0.8
– 0.6
– 0.4
– 0.2
0
χ
0
– 0.2
– 0.4
– 0.6
– 0.8
p


B
80
60
40
20
0
B (μT)
0.4
0.5
0.6
0.7
0.8
S/R ln2


8
6
4
2
0
0
2
4
6
8
|1/χ|
| T | (nK)


J_2/h (Hz)
20
AF1
J_1/h (Hz)
FM
20
−20
AF3
AF2
−20